ATMOSPHERIC TRACE MOLECULE SPECTROSCOPY

NASA-TM-108720

ATMOS

Long-term Atmospheric Measurement for Mission to Planet Earth

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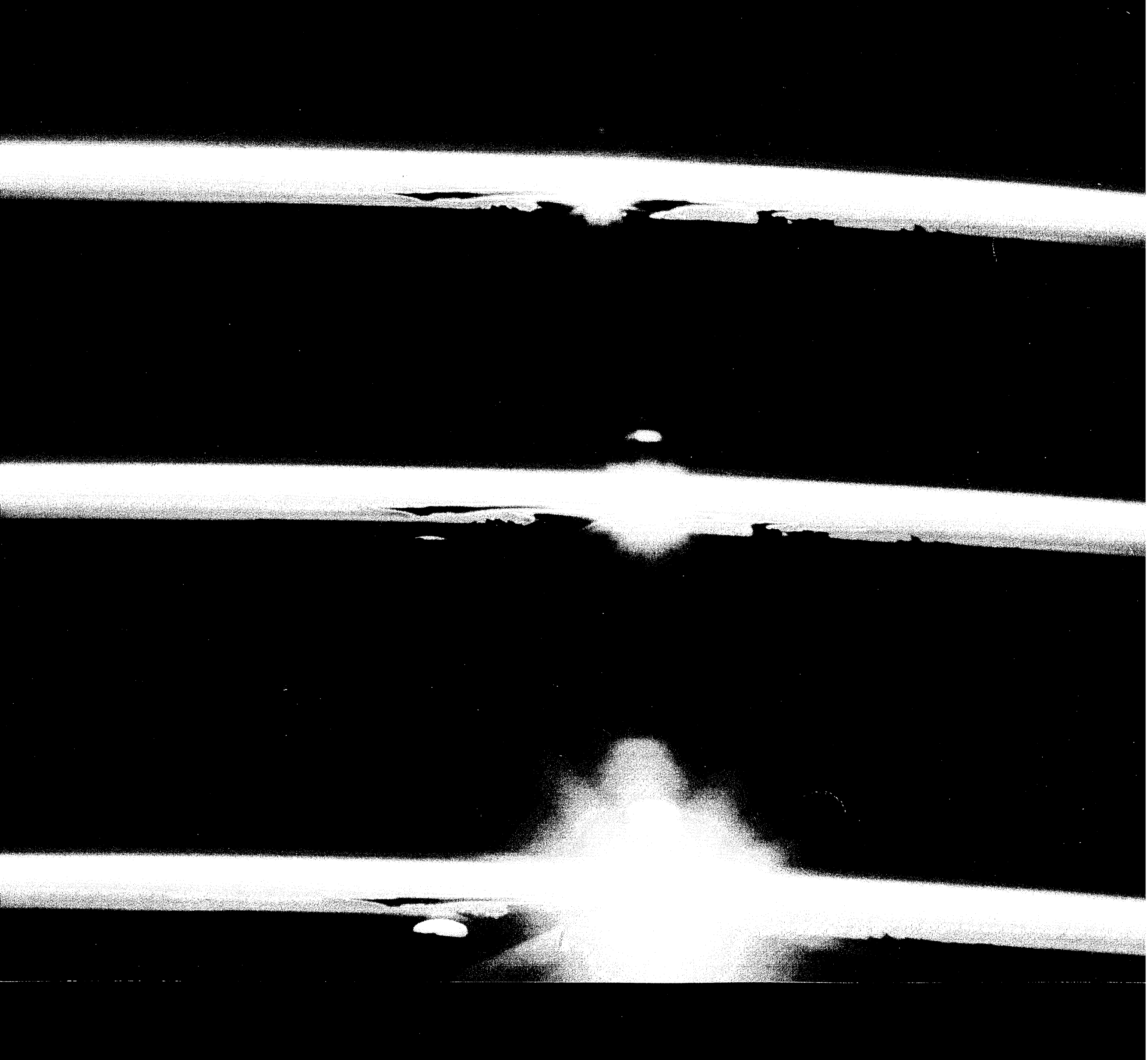

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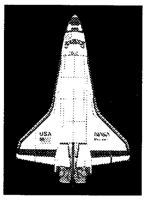
Long-term Atmospheric Measurements –Essential to Mission to Planet Earth



Over the last three or four decades scientists have achieved a major advance in our understanding of Earth's atmosphere. Until the 1950's, there were few reliable measurements of what the atmosphere itself was made up of, and as a consequence, there was no reason to think that it was anything other than a relatively stable and unchanging mass of air. Since then, instrumental and technological advances have led to much better measurements that reveal distinct regions of Earth's atmosphere–distinguished by their temperature structure and chemical composition *(see Figure 1)*. The composition within these regions is constantly evolving and is influenced both by natural changes and changes due to human activity.

Natural changes occur frequently. For example, seasonal changes result from changes in the amount of radiation reaching the atmosphere from the Sun over time. There are also day-night changes, for the same reason. More exotic influences include fluctuations in the solar wind and in cosmic rays from space, which also affect conditions in the atmosphere. Dramatic physical events such as volcanic eruptions inject dust, ash, and a variety of chemical compounds into the stratosphere.

According to data from satellites now in operation, the 1991 eruption of Mount Pinatubo, the largest volcanic eruption this century, has had a major impact on stratospheric chemistry by increasing the amount of sulfate aerosols (suspensions of solids or liquids in gases) in the lower stratosphere.

Today, most studies of the atmosphere focus on one of two interrelated phenomena resulting from human activity. One major concern is that of stratospheric ozone depletion caused by man-made chemicals such as chlorofluorocarbons (CFC's), used as refrigerants, cleaning solvents, and aerosol propellants. The ozone layer protects life on Earth from hazardous ultraviolet radiation from the Sun and so any weakening of that protective layer is a potential threat to Earth's life forms.

The second major concern is the increase in atmospheric concentration of gases such as carbon dioxide, methane, and nitrous oxide that at high concentrations are capable of trapping solar heat in the atmosphere, preventing it from being reradiated to space, and so potentially

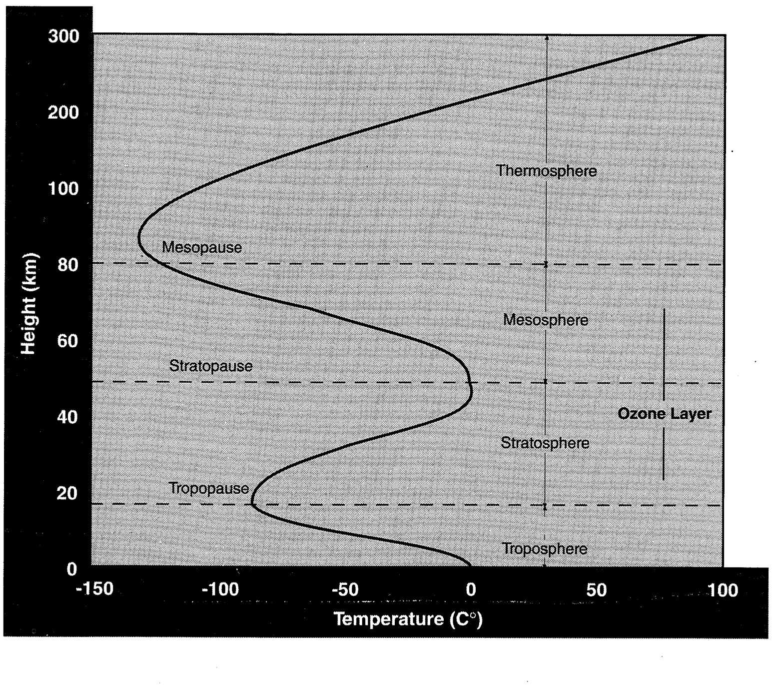


Figure 1: Graph showing the different regions of the Earth's atmosphere, delineated by the temperature variation with height.

Figure 2: Stratospheric ozone formation: ultraviolet radiation causes the dissociation of oxygen (O_2) molecules; the free oxygen atoms combine with other oxygen molecules to form ozone (O_3).

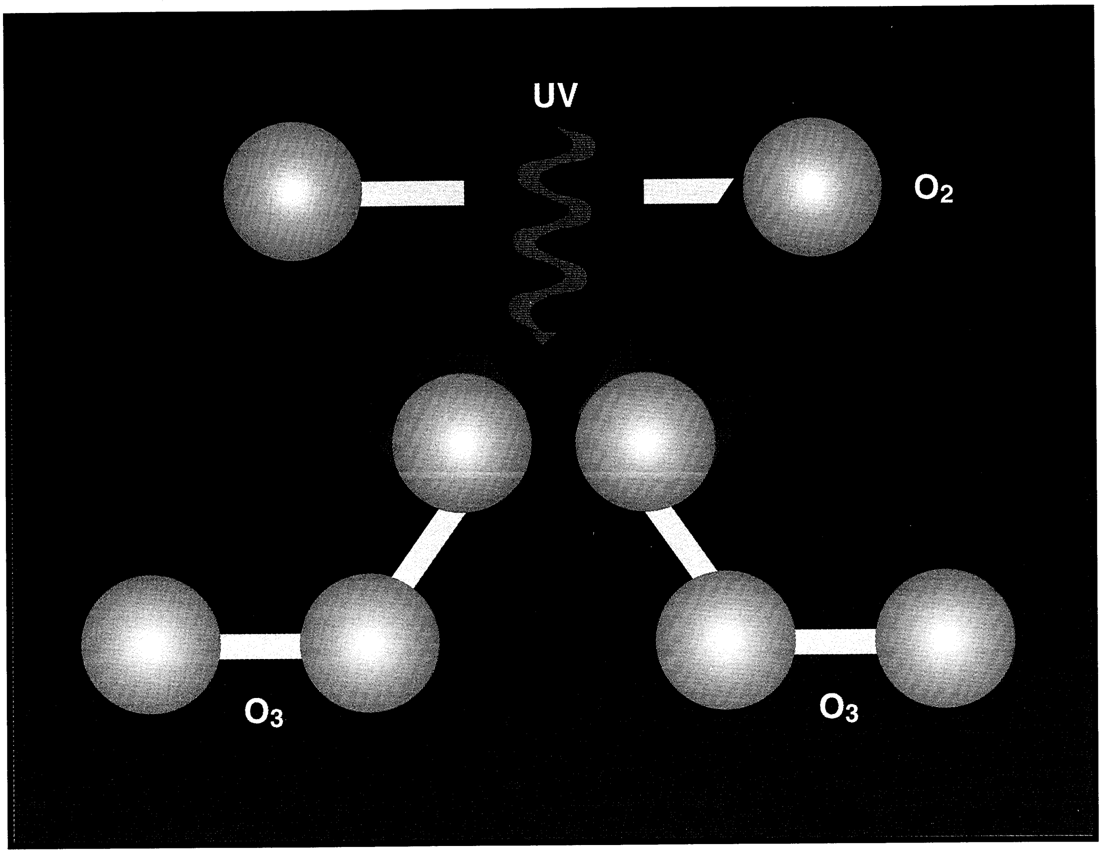
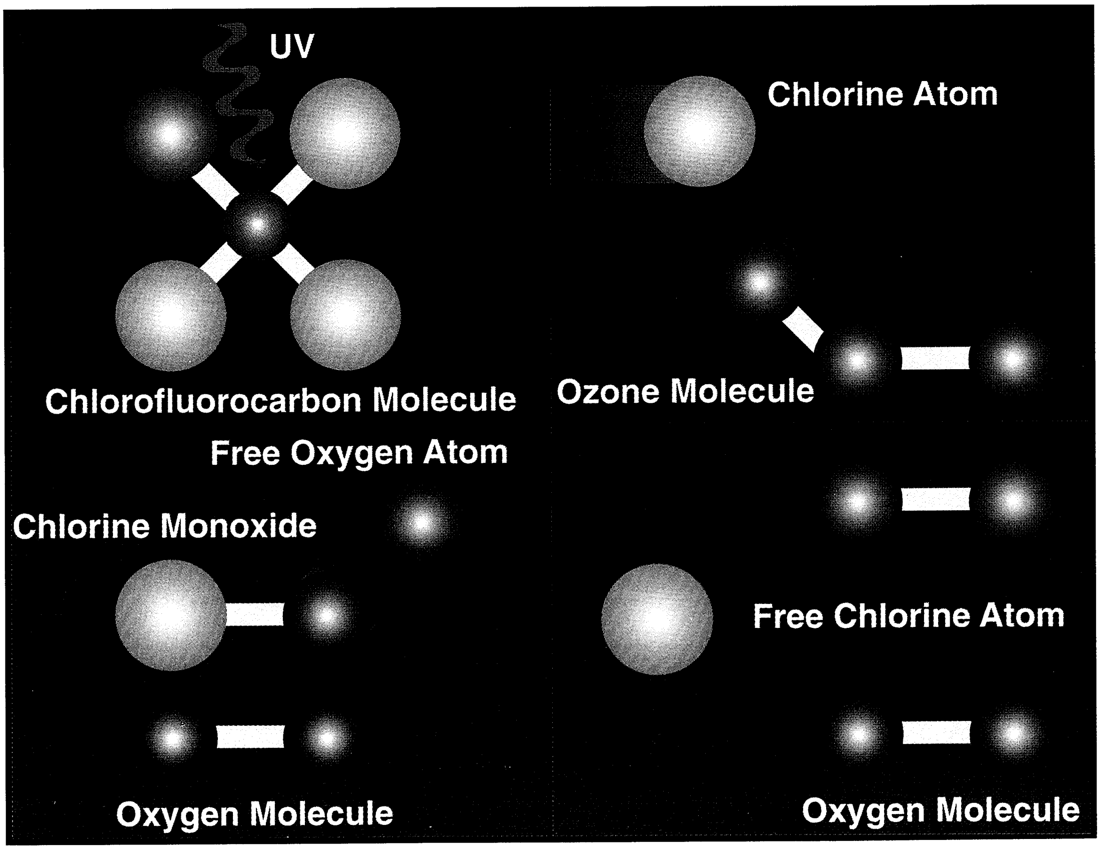


Figure 3: Stratospheric ozone destruction: chlorine released from CFC molecules destroys ozone in a catalytic reaction, surviving the process to go on to destroy other ozone molecules.



warming the planet–the "greenhouse effect." Researchers believe that increases of these gases–as a result, for example, of continuing world industrialization and more intensive farming–may eventually cause global average surface temperatures to rise significantly, with important consequences for agriculture, sea level, and weather patterns.

It has become clear to concerned scientists that a comprehensive study of Earth's atmosphere is necessary to establish the nature and significance of these changes in its composition and behavior. Because of the complexity and the dynamic nature of atmospheric chemical interactions, any attempt to understand these processes requires the simultaneous measurement of a large number of chemical species.

Two types of scientific investigations are required: global-scale studies of the trends of atmospheric change and more localized studies of the details of these changes, to serve as the foundation for future predictions. Space-based measurements can provide global-scale studies, since global coverage of the entire atmosphere can be obtained in a short period of time. More localized, detailed studies are best conducted with instruments flown on aircraft and balloon platforms from a number of locations around the globe. Scientists need these data as soon as possible if they are to understand the middle and upper atmosphere in sufficient detail to forecast the effects of present and future alterations in its composition.

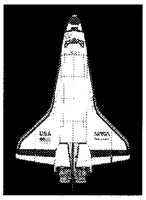
So a long-term, space-based measurement program, together with continued balloon and aircraft-borne investigations, is essential to monitor the predicted effects, to determine to what extent the concentration measurements agree with current models of stratospheric chemistry, and to determine the condition of the ozone layer. The Atmospheric Trace Molecule Spectroscopy (ATMOS) Experiment is currently making comprehensive, global measurements of Earth's atmosphere as part of the Atmospheric Laboratory for Applications and Science (ATLAS) program on the Space Shuttle. Part of NASA's Mission to Planet Earth, ATLAS is a continuing series of missions to study Earth and the Sun and provide a more fundamental understanding of the solar influences on Earth's atmosphere.

Chemistry in the Stratosphere

Short wavelength radiation from the Sun is absorbed in the Earth's upper atmosphere as it passes through the lower thermosphere, mesosphere, and stratosphere and is primarily responsible for the chemical reactions and dynamics of these regions. In the upper region of the stratosphere, ultraviolet light causes oxygen to dissociate and recombine to produce ozone, as shown in *Figure 2*.

The ozone layer shields life on Earth from the harmful effects of ultraviolet radiation. Under normal circumstances, the ozone balance is maintained by a range of competing natural chemical reactions. However, man-made chlorofluorocarbons released at the surface, eventually reaching this region, are broken down by sunlight, in a process called photolysis, to produce chlorine atoms. These atoms then destroy ozone in a catalytic cycle (i.e., one in which one of the reactants is left unaltered and free to react again) which is itself driven by solar radiation, as shown in *Figure 3*.

In addition to the chlorine family of gases, the nitrogen (N) and hydrogen (H) groups are also important in stratospheric chemistry. Each contains three basic types of species: source molecules, free radicals, and sink or reservoir molecules. Source species are relatively stable molecules which are dissociated either chemically or photolytically to produce the highly reactive free radicals which catalytically destroy ozone in a series of stratospheric chain reactions. Ultimately, the radicals are recombined into the inactive sink species, which on reaching the troposphere, are rained out of the atmosphere.



The ATMOS Program is designed primarily to measure the detailed composition of the stratosphere by detecting as many of its constituent gases as possible and determining how they are distributed vertically and horizontally. The ATMOS instrument detects variations in the amount of radiation from the Sun at infrared (IR) frequencies as the radiation passes through the atmosphere and is absorbed by the molecules present in its path. Records of these variations as a function of frequency are called absorption spectra, and the information they contain ultimately enables researchers to establish an important simultaneous inventory of upper atmospheric constituents.

The scientific objectives of the ATMOS program include the following:

• To provide an inventory of the constituent gases of the Earth's atmosphere.

• To determine how these gases are distributed in the atmosphere as a function of height, latitude, and longitude.

• To determine how the inventory changes with the seasons.

• To determine the lifetimes of gases in the atmosphere i.e. how long on average a gas molecule survives. This information is necessary for predicting the long-term consequences of current human activities which alter atmospheric composition.

• To study the effects of changes in solar output on the atmosphere.

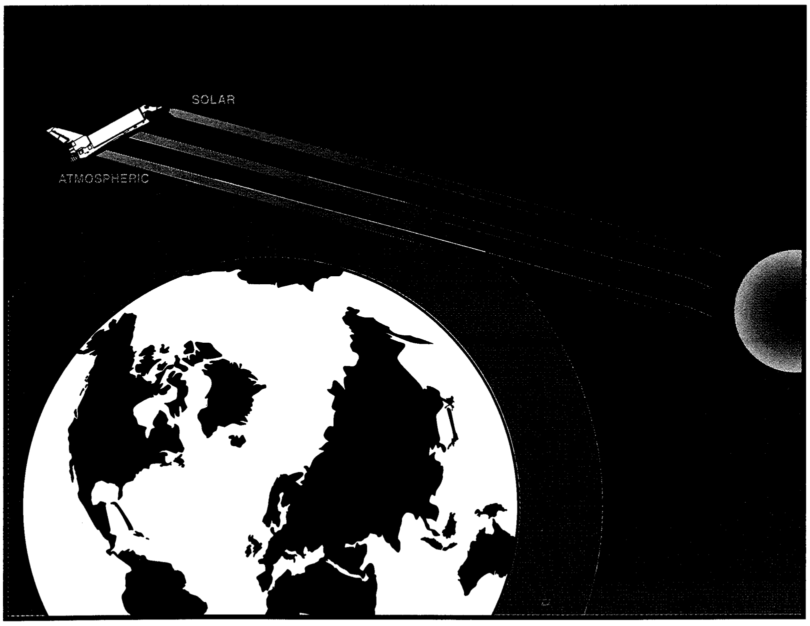


Figure 4: The limb-sounding technique from the Shuttle. The rays of light from the Sun to the spacecraft are called "line-of-sight" rays.

The ATMOS instrument uses a technique called limb sounding from the Shuttle to acquire information with regard to the concentrations and distributions of the gases present in the Earth's atmosphere; this technique is illustrated in *Figure 4*. The instrument detects solar IR radiation at wavelengths between 2.5 and 16 micrometers (μm) during the numerous sunsets and sunrises experienced from the orbiting Shuttle. Because the molecules of interest absorb radiation at very specific wavelengths in this infrared region, in amounts proportional to the number of molecules present, the absorption patterns in the spectra identify each species present as well as its concentration. Examples of the absorption patterns for a number of atmospheric molecules at different heights above the surface are shown in *Figure 5*.

The instrument makes successive measurements as the Sun either descends toward or rises above the Earth's surface with the Sun's rays passing through the atmosphere before reaching the instrument. As the Shuttle moves, this line of sight between the Sun and the instrument eventually is either blocked by the Earth or rises above the atmosphere. In a typical orbit, the height of the line-of-sight ray with respect to the Earth's surface during orbital sunsets and sunrises changes at a rate of about 1 to 2 kilometers per second; the ATMOS instrument was designed to record one complete spectrum every two seconds, and it thus obtains vertical distribution information of the upper atmospheric constituents with a resolution of 2 to 4 kilometers at all altitudes up to 140 kilometers.

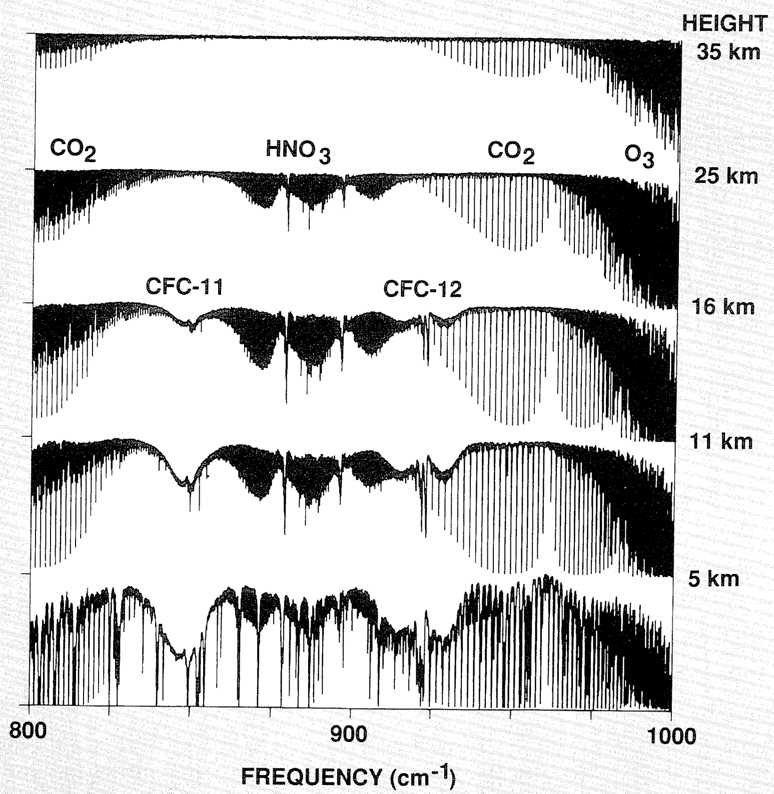


Figure 5: Absorption patterns of some atmospheric gases in spectra recorded at several heights above the Earth's surface. The frequency units, cm^{-1}, are equivalent to wavelength units of micrometers as the inverse x 10000. For example, 1000 cm^{-1} = 10 micrometers, and 800 cm^{-1} = 12.5 micrometers.

Why Simultaneous Measurements are Important

The simultaneity of the measurements of different gases is an important aspect of the experiment by virtue of the highly interactive nature of the photochemical processes that occur in the stratosphere. To modelers attempting to predict the effects of the chlorine cycle, for example, simultaneous measurements of as many chlorine compounds as possible, together with measurements of the natural minor and trace species, are of far greater benefit than indirectly related individual measurements of chlorine species taken at different times and locations. There is a great deal of natural variation in the concentrations of atmospheric gases, as a function of time, latitude, and season. In addition, different instruments which have not been cross-calibrated often produce different results, even when measurements are made simultaneously. Measurements of a number of related species made with a single instrument at the same time and place can help eliminate uncertainties in the relative concentrations of the gases being measured.

Figure 6: The ATMOS instrument.

ATMOS FTS Specifications

Configuration:
Cats' eye retroreflectors, double passed, continuous scan
Spectral Coverage:
2.1 to 16 micrometers (600 to $4800\ cm^{-1}$).
Resolution:
$0.01\ cm^{-1}$, unapodized
Optical Path Difference:
±50 centimeters
Path Difference Reference:
Stabilized He/Ne laser
Beamsplitter and Compensator:
Potassium bromide
Field of View:
1 to 4 mrad, selectable
Detector:
HgCdTe @ 77K.
Operating Temperature:
-5 to +45 degrees C.
Operating Power:
225 Watts DC, 135 Watts AC.
Data Rate:
16 Megabits per second
System Weight:
250 kilograms

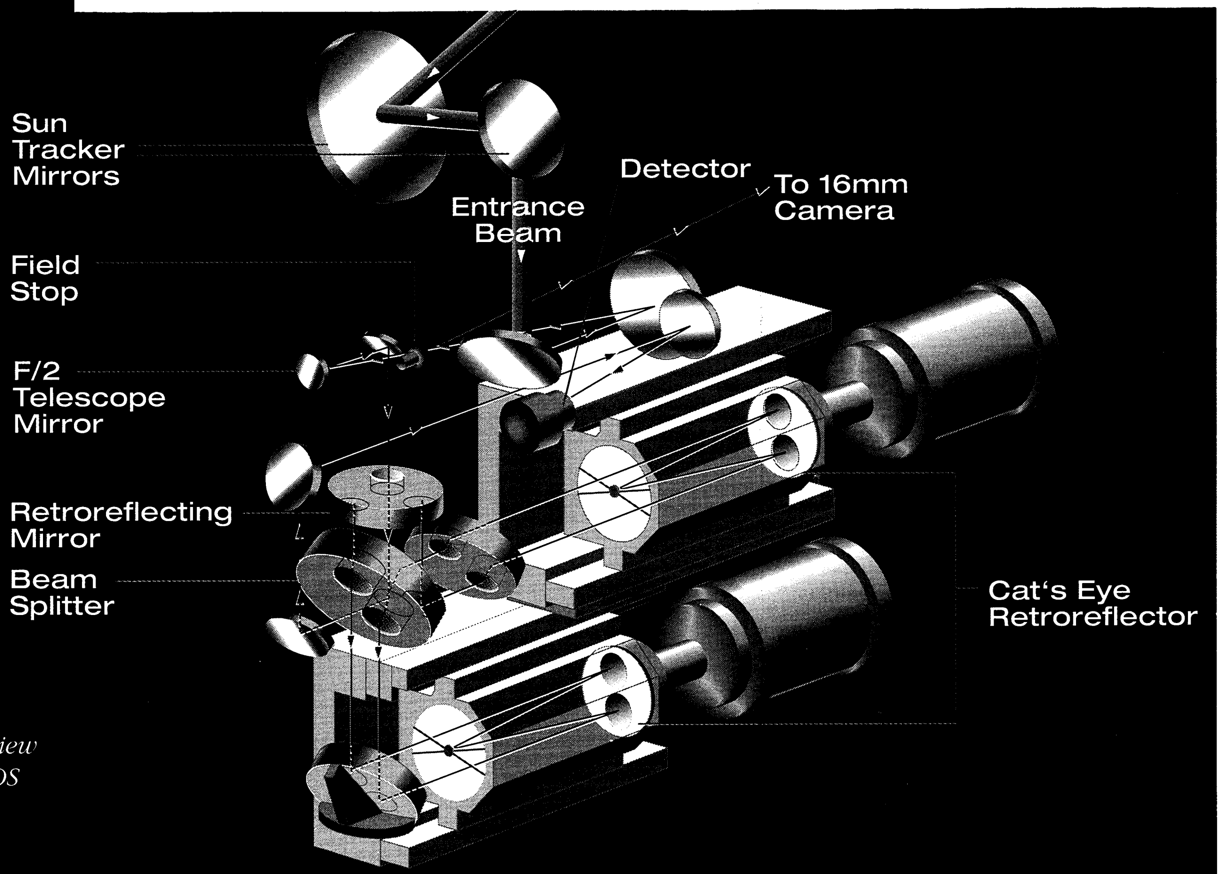
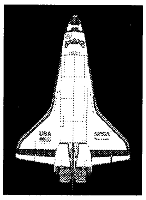


Figure 7: Exploded schematic view of the ATMOS instrument.



The ATMOS sensor, developed at NASA's Jet Propulsion Laboratory as part of a long-term atmospheric measurement program, is a state-of-the-art version *(see Figure 6)* of an instrument invented in the 19th century by Nobel-laureate physicist Albert Michelson from the University of Chicago. It uses the interference properties of light to determine the frequency characteristics of radiation passing through it by splitting the incoming light into two beams and then, by the use of moving mirrors, varying the distance one of the beams travels with respect to the other before they are recombined. Interference patterns are created that contain information on the frequencies and intensities of the light rays in the incoming light beam.

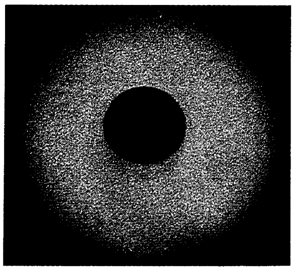


Figure 8: Frame camera image of the Sun with the ATMOS field of view (FOV) superimposed.

The major elements of the ATMOS instrument *(see Figure 7)* are a suntracker, a telescope, an interferometer, an IR detector, and a data processor. The suntracker keeps the field-of-view (FOV) of the instrument positioned on the Sun, and the telescope gathers the solar radiation for optical processing by the interferometer. To improve performance, the wavelength region covered by the instrument–near and middle infrared (IR) wavelengths from 2.5 to 16 μm–is divided into smaller intervals using selectable bandpass filters. During a data-gathering sequence, radiation exiting the interferometer is passed through one of these filters and is then focused onto a mercury-cadmium-tellurium (HgCdTe) detector, mechanically cooled to 77 Kelvin (K). The output from the detector is amplified, filtered and digitized and, during a normal mission, is then multiplexed into the downlink telemetry system or recorded on an on-board recorder. *Table 1* shows the species that can be identified using each optical filter.

The pointing capability of the suntracker provides nearly hemispheric coverage for acquiring the Sun at sunset or sunrise. In the tracking mode the FOV is centered on the Sun, but its position is commandable to any location on the disc. Radiation enters the instrument via the suntracker and passes to the telescope system, whch defines the instrument FOV and the size of the beam by the use of a very small selectable aperture. This aperture, through which the light must pass before it enters the interferometer, is located in the center of a metal disc. Light rejected by the disc is reflected to a 16mm frame camera that records, at the end of each full scan, an image of the Sun with the field-of-view superimposed on it *(Figure 8.)*

Table 1: Transmission Ranges of ATMOS Optical Filters.

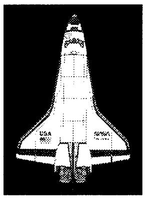
← — — — Band 5: 500-2450 cm^{-1} — — — →

Band 1 600-1200 cm^{-1}	Band 2 1100-2000 cm^{-1}	Band 3 1580-3400 cm^{-1}	Band 4 3100-4700 cm^{-1}
CO_2	CO_2	CO_2	CO_2
O_3	H_2O	H_2O	H_2O
	CH_4	CH_4	CH_4
HNO_3	N_2O	N_2O	N_2O
HNO_4	O_3	O_3	
N_2O_5	O_2	CO	HF
		N_2	
$ClNO_3$			HCN
CFC-11	HNO_3	NO	
CFC-12	NO	NO_2	
HCFC-22	NO_2		
CCl_4	N_2O_5	HCl	
COF_2		CH_3Cl	
	COF_2		
	CF_4		
C_2H_6		HCN	
C_2H_2		OCS	
SF_6		C_2H_6	

Legend:

O_2	molecular oxygen
O_3	ozone
CO	carbon monoxide
CO_2	carbon dioxide
COF_2	carbonyl difluoride
CH_4	methane
C_2H_2	acetylene
C_2H_6	ethane
Cl	chlorine
CCl_4	carbon tetrachloride
CCl_3F (CFC-11)	trichlorofluoromethane
CCl_2F_2 (CFC-12)	dichlorodifluoromethane
CF_4	carbon tetrafluoride
$ClONO_3$	chlorine nitrate
CH_3Cl	methyl chloride
$CHClF_2$ (HCFC-22)	chlorodifluoromethane
H_2O	water vapor
HNO_3	nitric acid
HNO_4	peroxynitric acid
HCN	hydrogen cyanide
H_2SO_4	sulfuric acid
HCl	hydrogen chloride
HF	hydrogen fluoride
N_2	molecular nitrogen
NO	nitric oxide
NO_2	nitrogen dioxide
N_2O	nitrous oxide
NO_x	oxides of nitrogen
N_2O_5	dinitrogen pentoxide
OCS	carbonyl sulfide
SF_6	sulfur hexafluoride

The Spacelab-3 Flight



ATMOS was flown for the first time from April 29 to May 5, 1985, as part of the science payload on board the Spacelab-3 Space Shuttle mission. On that first flight, the instrument detected the presence of some 40 different atmospheric constituents up to heights of 150 kilometers. The data from the flight produced the first simultaneous inventory of the majority of important stratospheric trace gases, including most of the molecules involved in ozone photochemistry *(Figure 9)*. The instrument observed a total of 20 sunset and sunrise events and acquired 1,192 atmospheric spectra and 1,474 direct solar spectra containing no atmospheric absorption features.

The geographic locations of the measurements are shown in *Figure 10*. The observed sunsets took place between the latitudes of 26 and 34 degrees North, and the sunrises near 48 degrees South latitude. A comparison of the results from individual sunset measurements suggests that there was little variability around the northern latitudes and longitudes sampled, with somewhat more variability apparent at southern latitudes. Part of this difference in variability might have been due to the differences in the seasons represented by the measurements; it was early spring in the northern hemisphere at the time of the measurements and early fall in the southern hemisphere.

The mission was also able to obtain important information regarding the air circulation patterns at play during the observation period. From an orbiting spacecraft, the observed frequency of the sunlight passing through the Earth's atmosphere changes as the spacecraft shifts from traveling toward the Sun at sunrise to going away from the Sun at sunset—much as a train's whistle shifts in frequency as the train roars by an observer. These frequency changes are measurable in the atmospheric spectrum. Atmospheric motions due to winds also contribute to these shifts, and by using this information it was possible to calculate the speed of zonal winds with a precision of some 2 meters per second throughout the stratosphere and mesosphere.

Some Principal Achievements of ATMOS on SL-3

- Provided self-contained ability to determine temperature, pressure, and molecular concentrations.
- Obtained concentration profiles for 25 different constituents in the altitude range from the surface to 140 km.
- Measured most of the NO_x family.
- Measured essentially complete inventory of halogen reservoir species.
- Measured HCl and HF to the top of the stratosphere (~60 km).
- Determined ozone chemistry in the mesosphere (O_3, H_2O, and temperature).
- Made initial detections or confirmed the presence of N_2O_5, HNO_4, COF_2, $ClONO_2$, CH_3Cl.
- Provided zonal wind profiles from 40 to 110 km.

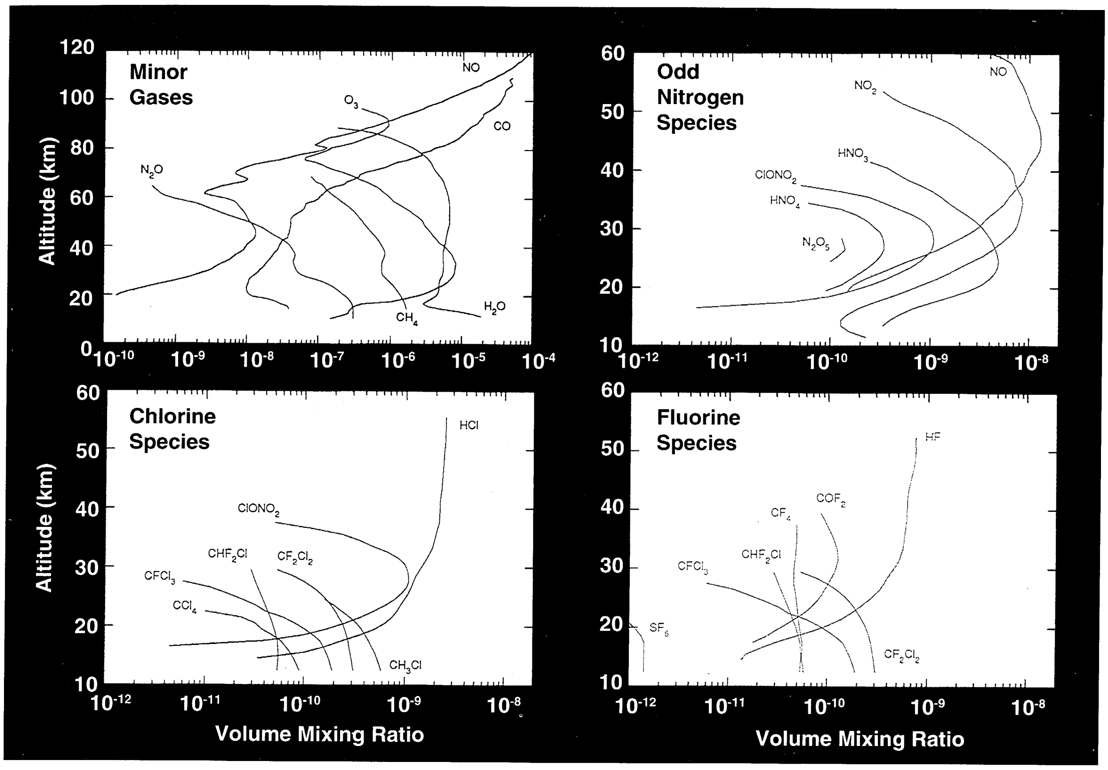


Figure 9: The vertical distributions of the gases measured by ATMOS in the northern hemisphere during Spacelab-3. The amount of gas is given by its volume, as a fraction of the volume of the atmosphere, and is called the "volume mixing ratio".

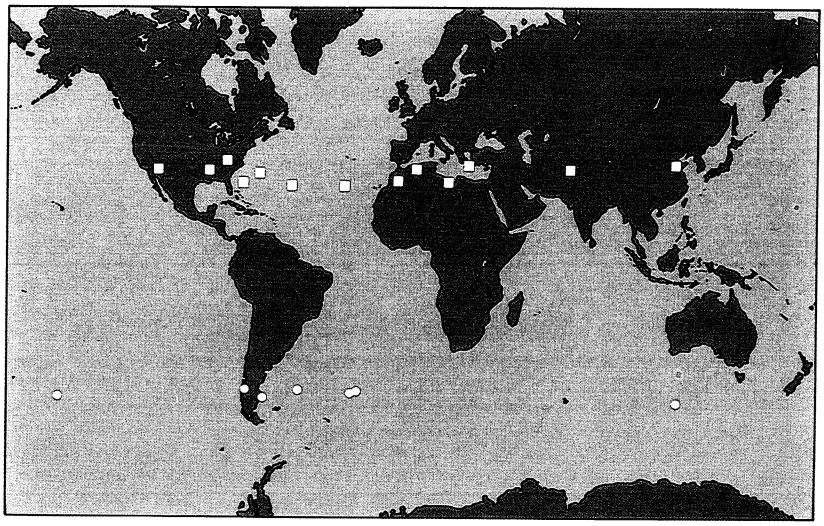


Figure 10: The geographic locations of the ATMOS Spacelab-3 atmospheric measurements. The squares represent sunsets, while the circles represent sunrises.

Solar Physics Results

A large amount of information of interest to solar physics has also been extracted from the data furnished by the Spacelab-3 flight of the ATMOS instrument.

A large number of the spectra returned by the experiment were purely solar,and this presented an opportunity to prepare a high-resolution, high signal-to-noise solar spectrum covering the entire 2.5 to 16 μm wavelength region of the infrared. These data have made possible the study of new molecular processes occurring at the very high temperatures near the Sun's surface and have permitted information to be derived on the composition and physical structure of the solar photosphere and chromosphere, two regions of the solar atmosphere. The abundance of carbon, nitrogen and oxygen in the Sun have been measured from Spacelab-3 data to help in determining the age of the solar system.

Figure 12: Comparison of two spectra taken by ATMOS at 18 km in 1985 and 1992. Note the presence in the 1992 spectrum of a large depression in the continuum caused by a sulfuric acid (H_2SO_4) aerosol feature as a result of material injected into the stratosphere during the Mt. Pinatubo eruption in 1991.

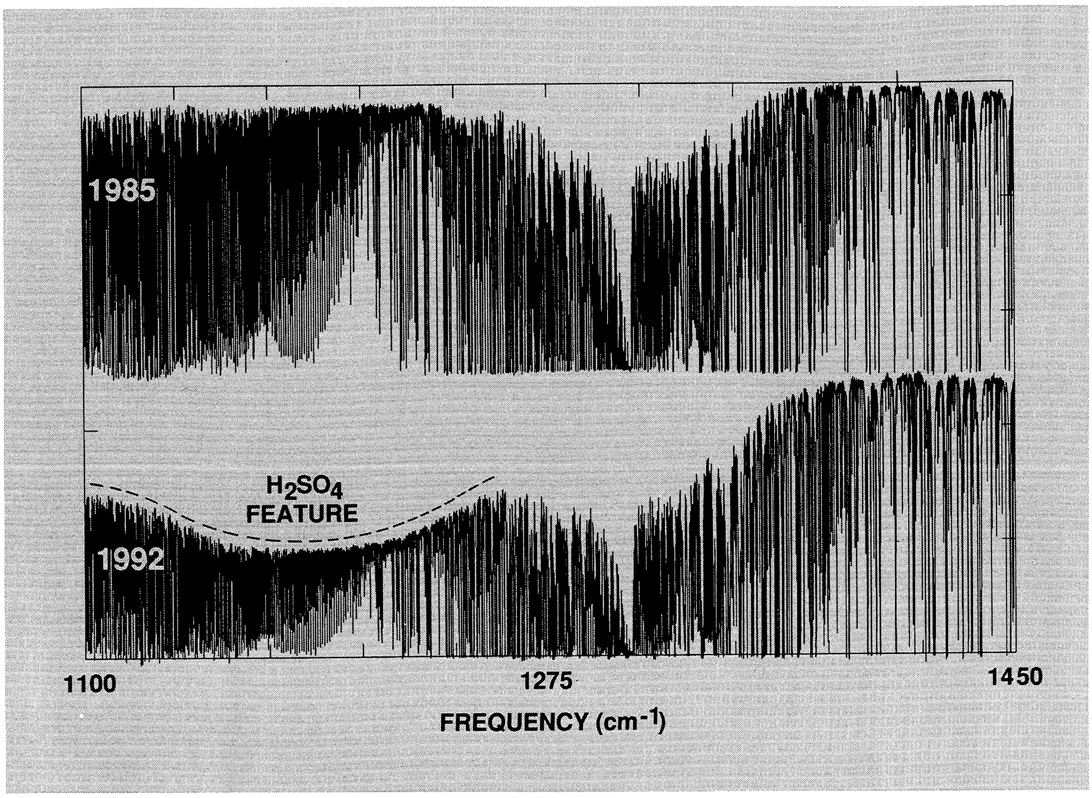
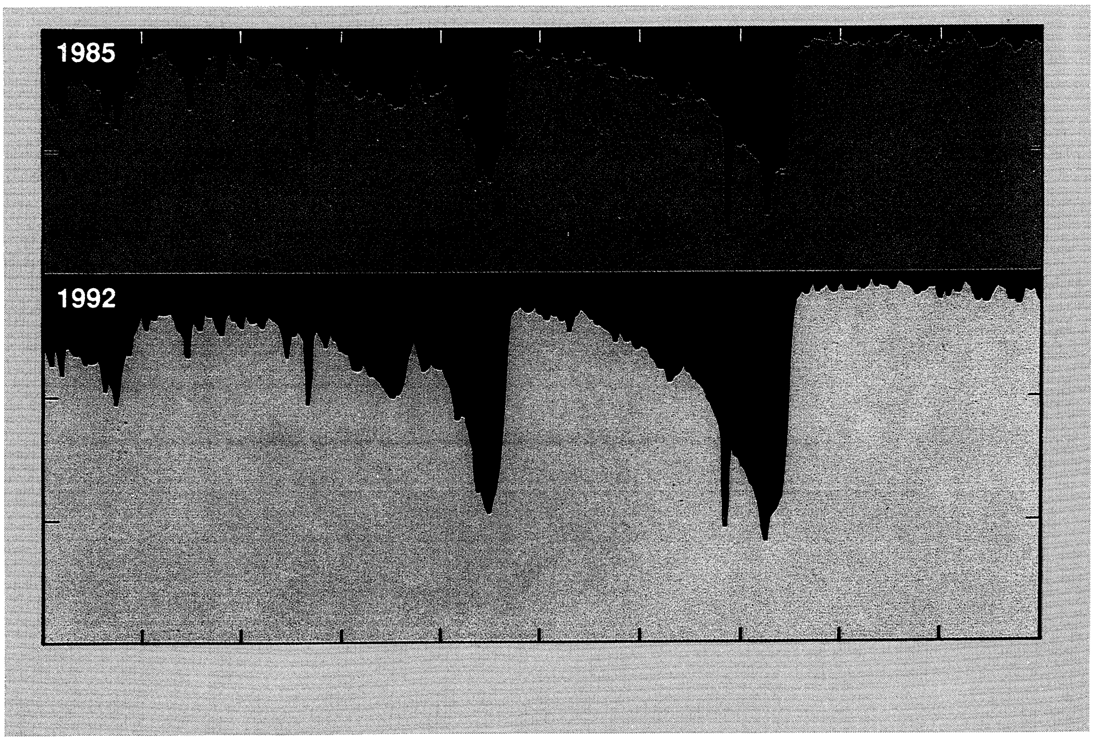
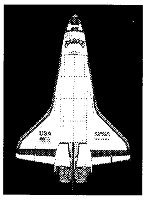


Figure 13: The increase in CFC amounts at 18 km observed by the ATMOS instrument from 1985 to 1992 is shown by the enlargement of the CFC features in the 1992 spectrum.



The Atlas-1 Flight



ATLAS-1, the first Atmospheric Laboratory for Applications and Science mission, flew on the Space Shuttle Atlantis from March 24 to April 2, 1992, with ATMOS among the 13 experiments on board. During the ATLAS-1 flight, ATMOS observed 99 sunsets and sunrises across latitudes from 55 degrees South to 30 degrees North *(Figure 11).* There were also three dedicated solar observations, taken on the solar limb rather than centered on the disk as in 1985, using ATMOS's field-of-view positioning capability. The ATLAS-1 data set was nearly ten times the size of that taken in the 1985 flight.

One of the unique results of the 1992 observations was the influence on the atmosphere of the Mount Pinatubo volcanic eruption in 1991. This was the largest volcanic eruption on the planet in this century, ejecting large quantities of sulfate aerosols into the stratosphere, detectable in the ATMOS data *(Figure 12).* The residual effects of this eruption are expected to remain for many years to come.

The science objectives for the investigation in 1992 included the following:

• To determine how the atmosphere has changed since 1985 due to both human activities and natural disturbances. The ATLAS-1 mission offered an unprecedented opportunity to observe the volcanically perturbed atmosphere by looking through the aerosol layer created by the eruption.

• To determine the latitudinal variation in atmospheric composition from the tropics to mid-latitudes. Because of circulation patterns, tropospheric air enters the stratosphere in the tropics and then moves toward the poles. The stratospheric air in mid-latitudes is thus "older" than tropical air, and by studying the atmosphere in different latitudinal regions, scientists can determine how long certain molecular species, for example the CFC's, exist at the higher atmospheric altitudes.

• To provide measurements for comparison with those being made by instruments on the Upper Atmosphere Research Satellite (UARS), launched in 1991 to study stratospheric change and ozone depletion.

• To determine to what extent the concentration measurements agree with current models of stratospheric chemistry, used for predicting future atmospheric changes.

Early analysis of the ATLAS-1 data set indicates that many of the types of changes which were predicted by the earlier chemical models of the stratosphere have occurred since the last ATMOS flight in 1985. Dramatic evidence for the increase in CFC's in the atmosphere is illustrated by the two spectra *(Figure 13)* which show the absorption due to these molecules in 1985 and later in 1992.

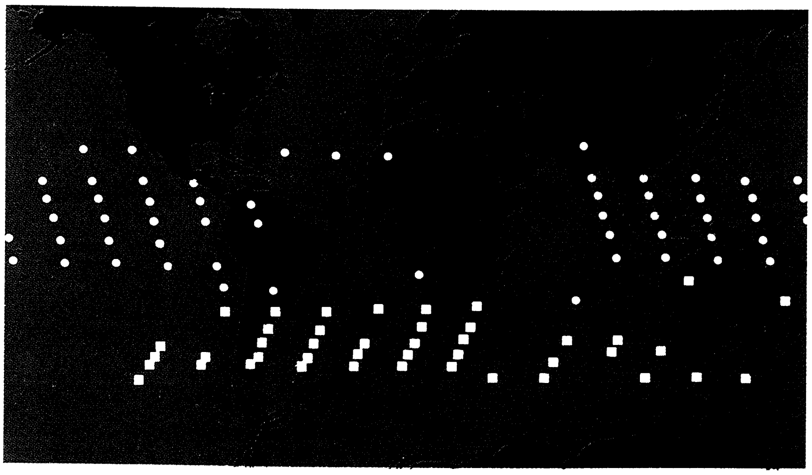


Figure 11: The geographic locations of the ATMOS atmospheric measurements from ATLAS-1.

ATMOS and other measurements and modeling programs have provided critical evidence that CFC's and other anthropogenic chemicals contribute to ozone depletion. As a result, with the signing of the Montreal Protocol in 1987 and its subsequent strengthening in 1990, world accord has been reached on phasing out the manufacture and use of CFC's in the industrialized countries. As these products are phased out, the task will still remain to monitor the long-term effects of those gases which have already been released into the atmosphere as they are slowly mixed (2 to 5 years) into the stratosphere and begin to interact with the ozone layer. Because of the slowness of the process, the residual effects of the CFC's will be present for many years into the future. ATMOS will continue to be part of the core payload of the ATLAS program with the flight of ATLAS-2 in 1993, and as such is expected to continue the monitoring efforts, which began with the Spacelab-3 flight, into the next century.

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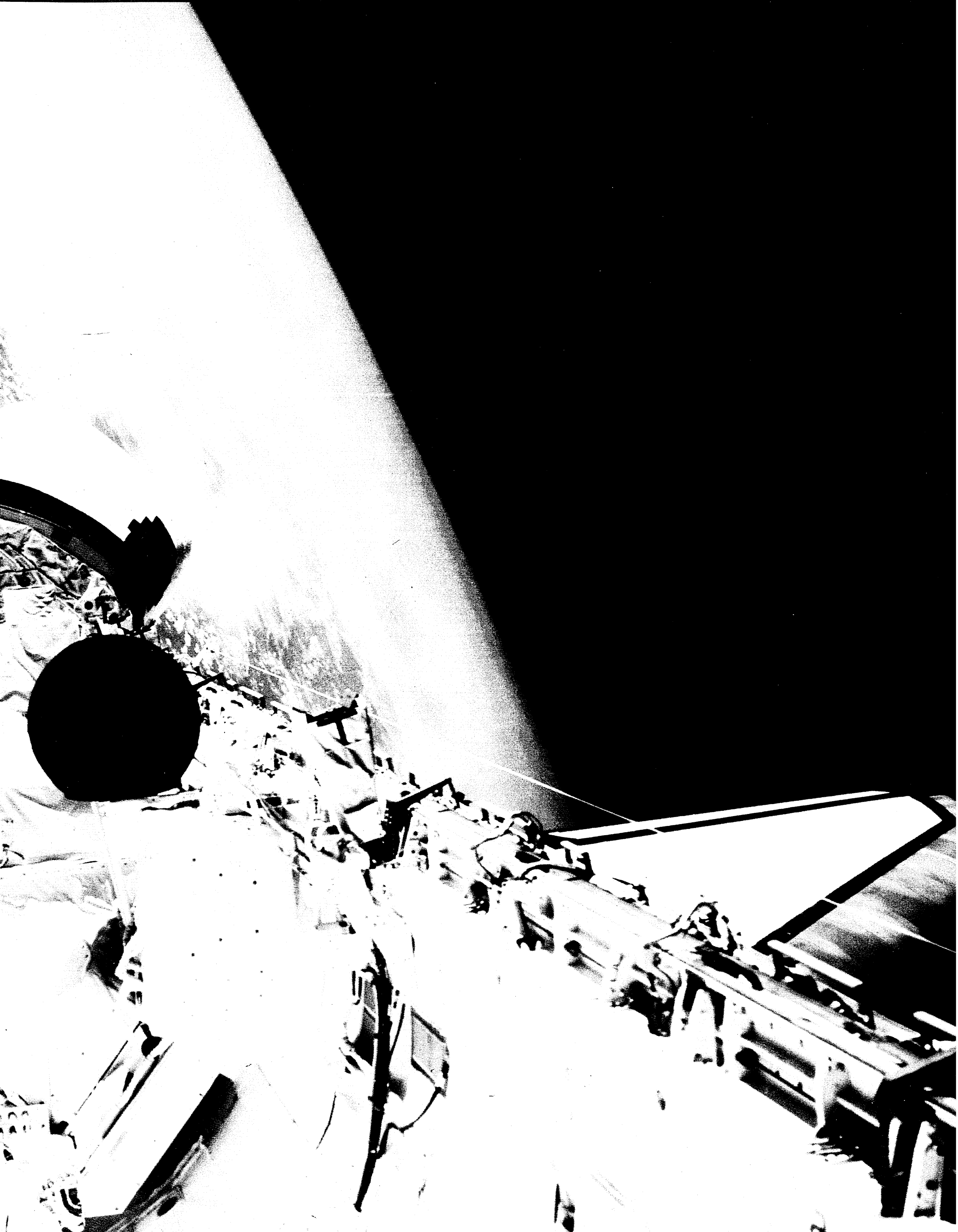

National Aeronautics and
Sapce Administration